

# Enhanced trap-assisted recombination in organic semiconductors

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## ABSTRACT

An analytical model to describe the interaction of excitons and charge transfer states with deep traps is formulated for the case of molecular materials. Here, we have considered the influence of a trap-assisted recombination on this phenomenon. The final expression for the effective recombination rate has been derived from the Shockley–Read–Hall theory and kinetic equations which characterize different photophysical processes. The presented model can be applied in modeling of organic photovoltaic devices.

## 1. Introduction

Over two last decades, electronics and photovoltaics based on organic materials have been extensively developed. Molecular devices can be flexible and their cost of fabrication is relatively low in comparison to the production of inorganic systems. However, there are still many problems with a recognition which physical mechanisms can improve functionality and stability of these devices.

Molecular excited states play a key role in the operation of organic systems. First, they can radiatively decay to ground states leading to a light emission [1]. Second, a generation of free charge carriers requires the dissociation of Frenkel-type excitons with involving of intermediate charge transfer (CT) states [2, 3, 4, 5]. Thus, a detailed investigation of the excited states effects is needed to better understand the photophysics of organic materials and, as a consequence, to construct more efficient devices.

An annihilation of excitons is one of several phenomena which cause the quenching of molecular excited states. Excitons can interact with other excitons or charge carriers [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21]. In the case of interaction with electrons and holes, a probability of such a process is greater when these carriers are located in trap states. Typically, the annihilation process is considered for carriers which occupy shallow traps. However, one can expect that molecular excitons or CT states can also be quenched by deep trap states.

The recombination is a main process which causes a decay of charge carriers. In organic materials, we predominantly observe a bimolecular recombination of the Langevin type [22]. However, it has been reported that also a non-Langevin recombination can appear in molecular systems and the origin of this anomalous effect has been widely discussed in the literature using different scenarios [23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41]. It seems that deep traps can visibly influence the recombination processes [42, 43]. Organic materials are usually considered as disordered systems with many trap states. Thus, it is reasonable to assume that a trap-assisted recombination via deep states, often called the Shockley–Read–Hall

(SRH) recombination [44, 45], can be partly responsible for the non-Langevin behavior.

The SRH recombination sometimes co-exists with other phenomena, such as the Auger recombination [46]. Recently, we have proposed that also an Auger-type decay of excitons [47] can occur together with a trap-assisted recombination [48]. The excitonic Auger process observed in inorganic materials may be approximately treated as an equivalent of the exciton annihilation by deep states in organic systems. However, the difference is that we can find more opportunities for molecular excited states because both interacting Frenkel molecular excitons and charge transfer states can be of singlet and triplet types. The aim of this paper is to formulate a theoretical model for a quenching of molecular excited states via deep traps in the presence of a trap-assisted recombination.

## 2. Model

For the typical SRH recombination mechanism [44, 45], there is an assumption that deep trap states are located in the middle of the gap. However, the SRH model can be sometimes applied in the case when traps lie also near band edges. Charge carriers are captured by trap states both from a conduction band (electrons) and from a valence band (holes). It should be noted that for molecular materials, we consider excited molecular states separately than CT states and free charge states (CS) [49, 50]. Thus, the conduction and the valence bands are associated with CS states for electrons ( $CS^{electrons}$ ) and CS states for holes ( $CS^{holes}$ ), respectively.

According to the SRH recombination, the capture rates for electrons ( $E_{cap}$ ) and holes ( $H_{cap}$ ) are defined as [44, 45]

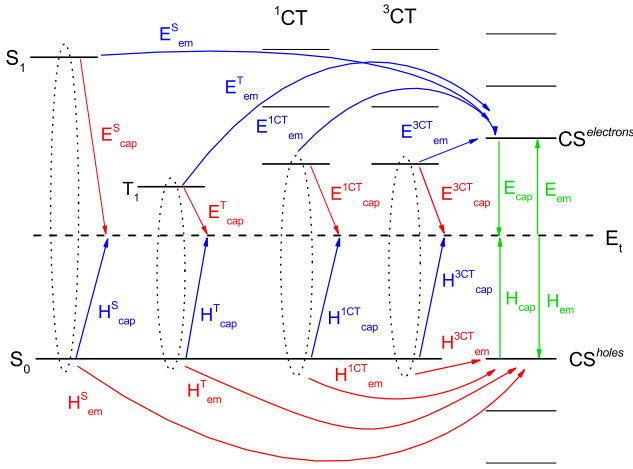
$$E_{cap} = C_n n N_t (1 - f_t) \quad (1)$$

and

$$H_{cap} = C_p p N_t f_t, \quad (2)$$

where  $C_n$  and  $C_p$  are capture coefficients for electrons and holes, respectively,  $n$  and  $p$  represent concentrations of electrons and holes in the conduction band and in the valence

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**Figure 1:** A diagram of the capture and emission processes considered in the model. Symbols  $S_0$ ,  $S_1$  and  $T_1$  are to denote a singlet ground state, a first excited singlet state and a first excited triplet state, respectively. Other symbols are explained in text and in Table 1. For each excitons and CT states, processes with the same colours (red or blue) have the same rate e.g. the rate for a capture of electrons from singlet excitons is equal to the rate of emission of holes from singlet excitons.

band, respectively,  $N_t$  is a parameter which defines a concentration of deep trap states and  $f_t$  denotes the probability that electrons occupy these states.

The emission rates for electrons ( $E_{em}$ ) and holes ( $H_{em}$ ) look as follows [44, 45]

$$E_{em} = C_n n_1 N_t f_t \quad (3)$$

and

$$H_{em} = C_p p_1 N_t (1 - f_t). \quad (4)$$

The concentrations  $n_1$  and  $p_1$  are given by

$$n_1 = N_c \exp\left(-\frac{E_c - E_t}{k_B T_{temp}}\right) \quad (5)$$

and

$$p_1 = N_v \exp\left(-\frac{E_t - E_v}{k_B T_{temp}}\right), \quad (6)$$

where  $N_c$  and  $N_v$  denote effective densities of states in the conduction band and in the valence band, respectively,  $E_c$  and  $E_v$  represent energies for the bottom level of the conduction band and for the top level of the valence band, respectively,  $E_t$  is a value of the trap states energy, whereas  $k_B$  and  $T_{temp}$  are the Boltzmann constant and the temperature, respectively.

Fig. 1 illustrates all capture and emission processes presented in the enhanced model. It shows that the interaction of an exciton or a CT state with a deep trap causes a simultaneous capture of a charge carrier from the interacting object and an emission of an opposite sign charge carrier to the CS

band. This is a reason why we will compare the capture rate for one carrier to the emission rate of an opposite carrier in the following equations (7)–(14) and we will not treat both processes as independent effects [48].

Now, we can consider an interaction of excitons with deep states. Similarly to the SRH formalism, the rates for a capture of electrons from singlet excitons ( $E_{cap}^S$ ) and from triplet excitons ( $E_{cap}^T$ ) may be written as

$$E_{cap}^S = A_{S_n} S N_t (1 - f_t) = H_{em}^S \quad (7)$$

and

$$E_{cap}^T = A_{T_n} T N_t (1 - f_t) = H_{em}^T, \quad (8)$$

where  $A_{S_n}$  and  $A_{T_n}$  represent capture coefficients for electrons from singlet and triplet excitons, respectively,  $S$  and  $T$  are concentrations of singlet and triplet excitons, respectively, whereas  $H_{em}^S$  and  $H_{em}^T$  denote the rates which characterize an emission of holes from singlet and triplet excitons, respectively.

The rates to describe a process of holes capture from singlet excitons ( $H_{cap}^S$ ) and from triplet excitons ( $H_{cap}^T$ ) can be expressed as

$$H_{cap}^S = A_{S_p} S N_t f_t = E_{em}^S \quad (9)$$

and

$$H_{cap}^T = A_{T_p} T N_t f_t = E_{em}^T, \quad (10)$$

where  $A_{S_p}$  and  $A_{T_p}$  denote capture coefficients for holes from singlet and triplet excitons, respectively, whereas  $E_{em}^S$  and  $E_{em}^T$  are the rates for electrons emission from singlet and triplet excitons, respectively.

Analogically, we can take into account an interaction of CT states with deep traps. It should be noted that a behavior of singlet charge transfer states ( $^1CT$ ) and triplet charge transfer states ( $^3CT$ ) should also be considered separately. The rates for a capture of electrons from  $^1CT$  states ( $E_{cap}^{1CT}$ ) and from  $^3CT$  states ( $E_{cap}^{3CT}$ ) can be defined as

$$E_{cap}^{1CT} = B_{1CT,n} X_{1CT} N_t (1 - f_t) = H_{em}^{1CT} \quad (11)$$

and

$$E_{cap}^{3CT} = B_{3CT,n} X_{3CT} N_t (1 - f_t) = H_{em}^{3CT}, \quad (12)$$

where  $B_{1CT,n}$  and  $B_{3CT,n}$  are capture coefficients for electrons from  $^1CT$  and  $^3CT$  states, respectively,  $X_{1CT}$  and  $X_{3CT}$  denote concentrations of  $^1CT$  and  $^3CT$  states, respectively, whereas  $H_{em}^{1CT}$  and  $H_{em}^{3CT}$  represent the rates which describe an emission of holes from  $^1CT$  and  $^3CT$  states, respectively.

The rates for a capture of holes from  $^1CT$  states ( $H_{cap}^{1CT}$ ) and from  $^3CT$  states ( $H_{cap}^{3CT}$ ) may be written as follows

$$H_{cap}^{1CT} = B_{1CT,p} X_{1CT} N_t f_t = E_{em}^{1CT} \quad (13)$$

and

$$H_{cap}^{3CT} = B_{3CT,p} X_{3CT} N_t f_t = E_{em}^{3CT}, \quad (14)$$

where  $B_{1CT,p}$  and  $B_{3CT,p}$  are capture coefficients for holes from  $^1CT$  and  $^3CT$  states, respectively, whereas  $E_{em}^{1CT}$  and  $E_{em}^{3CT}$  denote the rates for electrons emission from  $^1CT$  and  $^3CT$  states, respectively.

Photons can excite molecules to higher singlet states, whereas means that all derivatives with respect to  $t$  are equal to zero in all kinetic equations (15)–(20). However, to determine the effective recombination rate via deep states for charge carriers ( $U$ ), it is enough to consider only equations (19) and (20), which describe population of electrons and holes in the bands. Thus, one has

$$\frac{\partial S}{\partial t} = G_{S,phot} + G_{S,1CT} - G_{T,isc} - F_{1CT,S} - R_S - E_{cap}^S - H_{cap}^S \quad (15)$$

and

$$\frac{\partial T}{\partial t} = G_{T,isc} + G_{T,3CT} - F_{3CT,T} - R_T - E_{cap}^T - H_{cap}^T, \quad (16)$$

where  $t$  is the time,  $G_{S,phot}$  denotes a generation rate of singlet excitons by photons,  $G_{T,isc}$  is a rate of triplet excitons generation via inter-system crossing,  $R_S$  and  $R_T$  represent monomolecular recombination rates of singlet and triplet excitons, respectively,  $G_{S,1CT}$  and  $G_{T,3CT}$  are generation rates of singlet excitons from  $^1CT$  states and triplet excitons from  $^3CT$  states, respectively, whereas  $F_{1CT,S}$  and  $F_{3CT,T}$  represent formation rates of  $^1CT$  states from singlet excitons and  $^3CT$  states from triplet excitons, respectively.

Similarly, we can write kinetic equations for the population of CT states and charge carriers

$$\frac{\partial X_{1CT}}{\partial t} = F_{1CT,S} + F_{1CT} - D_{1CT} - G_{S,1CT} - F_{3CT,isc} - R_{1CT} - E_{cap}^{1CT} - H_{cap}^{1CT}, \quad (17)$$

$$\frac{\partial X_{3CT}}{\partial t} = F_{3CT,T} + F_{3CT} - D_{3CT} - G_{T,3CT} + F_{3CT,isc} - R_{3CT} - E_{cap}^{3CT} - H_{cap}^{3CT}, \quad (18)$$

$$\frac{\partial n}{\partial t} = D_{1CT} + D_{3CT} - F_{1CT} - F_{3CT} - E_{cap} + E_{em} + E_{em}^S + E_{em}^T + E_{em}^{1CT} + E_{em}^{3CT} \quad (19)$$

and

$$\frac{\partial p}{\partial t} = D_{1CT} + D_{3CT} - F_{1CT} - F_{3CT} - H_{cap} + H_{em} + H_{em}^S + H_{em}^T + H_{em}^{1CT} + H_{em}^{3CT} \quad (20)$$

where  $F_{1CT}$  and  $F_{3CT}$  are formation rates of  $^1CT$  and  $^3CT$  states from free charge carriers, respectively,  $F_{3CT,isc}$  represents a rate of  $^3CT$  states formation via inter-system crossing,  $D_{1CT}$  and  $D_{3CT}$  are dissociation rates of  $^1CT$  and  $^3CT$  states into separated charge carriers, respectively, whereas

$R_{1CT}$  and  $R_{3CT}$  denote monomolecular recombination rates of  $^1CT$  and  $^3CT$  states, respectively.

It should be noted that, for a clarity, the mentioned above processes of generation, formation and dissociation are not presented in Fig. 1. However, they do not influence on the final recombination rate which will be presented in the following, see equation (21).

Now, we can take into account steady state conditions. It means that all derivatives with respect to  $t$  are equal to zero in all kinetic equations (15)–(20). However, to determine the effective recombination rate via deep states for charge carriers ( $U$ ), it is enough to consider only equations (19) and (20), which describe population of electrons and holes in the bands. Thus, one has

$$U = E_{cap} - E_{em} - E_{em}^S - E_{em}^T - E_{em}^{1CT} - E_{em}^{3CT} = H_{cap} - H_{em} - H_{em}^S - H_{em}^T - H_{em}^{1CT} - H_{em}^{3CT}. \quad (21)$$

Next, the trap occupancy fractions may be found in the forms

$$f_t = \left( C_n n + C_p p_1 + \sum_{Y \in \{S,T\}} A_{Yn} Y + \sum_{i \in \{1CT,3CT\}} B_{i,n} X_i \right) \times \left[ \sum_{q \in \{n,p\}} C_q (q + q_1) + \sum_{Y \in \{S,T\}} (A_{Yn} + A_{Yp}) Y + \sum_{i \in \{1CT,3CT\}} (B_{i,n} + B_{i,p}) X_i \right]^{-1} \quad (22)$$

for electrons and

$$1 - f_t = \left( C_n n_1 + C_p p + \sum_{Y \in \{S,T\}} A_{Yp} Y + \sum_{i \in \{1CT,3CT\}} B_{i,p} X_i \right) \times \left[ \sum_{q \in \{n,p\}} C_q (q + q_1) + \sum_{Y \in \{S,T\}} (A_{Yn} + A_{Yp}) Y + \sum_{i \in \{1CT,3CT\}} (B_{i,n} + B_{i,p}) X_i \right]^{-1} \quad (23)$$

for holes.

With the use of these formulas, we can derive the following expression for the effective recombination rate

$$U = \frac{n^{(EF)} p^{(EF)} - n_{int}^2}{\tau_p^{(EF)} (n^{(EF)} + n_1) + \tau_n^{(EF)} (p^{(EF)} + p_1)}, \quad (24)$$

where

$$n_{int} = (N_c N_v)^{1/2} \exp \left( - \frac{\Delta E}{2k_B T_{temp}} \right) \quad (25)$$

is an intrinsic concentration of charge carriers with the parameter  $\Delta E$  which represents an energy of a band gap.

We can see that equation (24) has a similar form to the original expression known from the SRH theory of recombination. However, the obtained parameters  $n^{(EF)}$ ,  $p^{(EF)}$ ,  $\tau_n^{(EF)}$  and  $\tau_p^{(EF)}$  have a different meaning.

The effective concentrations may be defined as

$$n^{(EF)} = \frac{C_n n_1 n}{C_n n_1 + \sum_{Y \in \{S,T\}} A_{Yp} Y + \sum_{i \in \{1CT,3CT\}} B_{i,p} X_i} \quad (26)$$

for electrons and

$$p^{(EF)} = \frac{C_p p_1 p}{C_p p_1 + \sum_{Y \in \{S,T\}} A_{Yn} Y + \sum_{i \in \{1CT,3CT\}} B_{i,n} X_i} \quad (27)$$

for holes, whereas the effective lifetimes for both types of charge carriers can be written in the forms

$$\tau_n^{(EF)} = \frac{n_1}{N_t \left( C_n n_1 + \sum_{Y \in \{S,T\}} A_{Yp} Y + \sum_{i \in \{1CT,3CT\}} B_{i,p} X_i \right)} \quad (28)$$

and

$$\tau_p^{(EF)} = \frac{p_1}{N_t \left( C_p p_1 + \sum_{Y \in \{S,T\}} A_{Yn} Y + \sum_{i \in \{1CT,3CT\}} B_{i,n} X_i \right)} \quad (29)$$

It is visible that all effective parameters strongly depend on the quenching of excitons and charge transfer states via deep traps. The quenching effect causes that  $n^{(EF)} < n$ ,  $p^{(EF)} < p$ ,  $\tau_n^{(EF)} < \tau_n$  and  $\tau_p^{(EF)} < \tau_p$ , where  $\tau_n = 1/(C_n N_t)$  and  $\tau_p = 1/(C_p N_t)$  represent the SRH lifetimes of electrons and holes, respectively. These SRH lifetimes are explicitly independent on the concentrations of charge carriers which are the only objects interacting with deep states in the SRH recombination. However, the derived effective lifetimes  $\tau_n^{(EF)}$  and  $\tau_p^{(EF)}$  depend on the concentration of interacting species (here, excitons and CT states).

The next conclusion is that the effective recombination rate  $U$  is lower than the rate obtained from the SRH theory. It can be intuitively found from equation (21) which is a mathematical equivalent of equation (24). When we neglect an influence of excitons and CT states, then we obtain  $U = E_{cap} - E_{em} = H_{cap} - H_{em}$  which can be treated as the other form of the SRH recombination rate. Thus, it is seen that an interaction of molecular exciton or CT state with deep states should decrease the value of  $U$ .

### 3. Summary

The quenching of singlet and triplet excitons and charge transfer states via deep traps has been taken into account for organic semiconductors in the case when a trap-assisted recombination takes place. In this work, we have derived the expression which describes the effective recombination

rate. The considered quenching processes can occur in photovoltaic devices based on molecular materials. Therefore, the presented model may be useful to simulate photophysical effects in organic photocells.

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**Table 1**

The capture and emission processes considered in the model.

Symbol	Process	Concentration	Coefficient
$E_{cap}$	capture of electrons from the CS conduction band ( $CS^{electrons}$ )	$n$	$C_n$
$H_{cap}$	capture of holes from the CS valence band ( $CS^{holes}$ )	$p$	$C_p$
$E_{em}$	emission of electrons from the trap states ( $E_t$ )	$n_1$	$C_n$
$H_{em}$	emission of holes from the trap states ( $E_t$ )	$p_1$	$C_p$
$E_{cap}^S$	capture of electrons from singlet excitons	$S$	$A_{Sn}$
$E_{cap}^T$	capture of electrons from triplet excitons	$T$	$A_{Tn}$
$H_{cap}^S$	capture of holes from singlet excitons	$S$	$A_{Sp}$
$H_{cap}^T$	capture of holes from triplet excitons	$T$	$A_{Tp}$
$E_{em}^S$	emission of electrons from singlet excitons	$S$	$A_{Sn}$
$E_{em}^T$	emission of electrons from triplet excitons	$T$	$A_{Tn}$
$H_{em}^S$	emission of holes from singlet excitons	$S$	$A_{Sp}$
$H_{em}^T$	emission of holes from triplet excitons	$T$	$A_{Tp}$
$E_{cap}^{1CT}$	capture of electrons from singlet CT states ( $^1CT$ )	$X_{1CT}$	$B_{1CT,n}$
$E_{cap}^{3CT}$	capture of electrons from triplet CT states ( $^3CT$ )	$X_{3CT}$	$B_{3CT,n}$
$H_{cap}^{1CT}$	capture of holes from singlet CT states ( $^1CT$ )	$X_{1CT}$	$B_{1CT,p}$
$H_{cap}^{3CT}$	capture of holes from triplet CT states ( $^3CT$ )	$X_{3CT}$	$B_{3CT,p}$
$E_{em}^{1CT}$	emission of electrons from singlet CT states ( $^1CT$ )	$X_{1CT}$	$B_{1CT,p}$
$E_{em}^{3CT}$	emission of electrons from triplet CT states ( $^3CT$ )	$X_{3CT}$	$B_{3CT,p}$
$H_{em}^{1CT}$	emission of holes from singlet CT states ( $^1CT$ )	$X_{1CT}$	$B_{1CT,n}$
$H_{em}^{3CT}$	emission of holes from triplet CT states ( $^3CT$ )	$X_{3CT}$	$B_{3CT,n}$